

Rabi Waves in Carbon Nanotubes - Experiment

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Rabi waves have been experimentally registered for the first time by Raman scattering studies of zigzag nanotubes, produced by high energy ion beam modification of natural diamond single crystals. Antiferroelectric spin wave resonance has been detected for the first time in Raman spectroscopy practice in given samples. Substantial qualitative and quantitative changes in Raman spectra in dependence on propagation direction of laser excitation wave have been found.

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The relativistic theory of quantized fields, in particular, quantum electrodynamics (QED) takes on more and more significance for its practical application and it, in fact, becomes to be working instrument in spectroscopy studies and industrial spectroscopy control. Presented work confirms given conclusion. The authors of recent publications [1], [2], in which the generalization of multi-qubit QED-model of [3] has been proposed by taking into account the interaction between the qubits and by inclusion in Hamiltonian of nonlinear term, have also predicted and theoretically studied the new coherent effect of nonlinear quantum optics – spatial propagation of Rabi oscillations - Rabi waves - in one-dimensional (1D) quantum dot chain. The similar effect is predicted in [4] for interacting multichain system, on the example of zigzag nanotubes (NT), in which the chains, being to be connected each other, are naturally strongly interact between themselves. The aim of given letter is experimental confirmation of the conclusions of the works [1], [2], [4], that is experimental evidence for the existence of Rabi wave phenomenon. Samples of type IIa natural diamond (nitrogen content was less than $5 \times 10^{17} \text{ cm}^{-3}$), implanted by high energy ions of copper (63 MeV, $5 \times 10^{14} \text{ cm}^{-2}$) and boron (13,6 MeV) have been studied. Ion implantation was performed along $\langle 111 \rangle$ crystal direction. Raman scattering (RS) spectra were registered in backscattering geometry. Laser excitation wave length was 488 nm, rectangular slit $350 \times 350 (\mu\text{m})^2$ was used, scan velocity was 100 cm^{-1} pro minute. The spectra observed are presented in Figures 1 to 3. We see from comparison of the spectra, presented in Figures 1 and 2, that they are strongly dependent on laser beam direction. The spectrum, presented in Figure 1, is characteristic of the only ion beam modified region of the sample, since characteristic diamond line near 1332 cm^{-1} is absent in the spectrum. The RS-lines with peak positions $656.8 \pm 0.2 \text{ cm}^{-1}$, $1215 \pm 1 \text{ cm}^{-1}$, $1779.5 \pm 1 \text{ cm}^{-1}$ and $2022.3 \pm 0.5 \text{ cm}^{-1}$ correspond to given region by laser excitation transversely to sample surface from implanted side. At the same time by laser excitation of the same sample from opposite unimplanted side the qualitatively other picture is observed, see Figure 2. Firstly, rather intensive relatively narrow line, which is characteristic for diamond single crystals, is now nat-

urally presented in the spectrum (full amplitude of given line is not shown in Figure 2). It is interesting, that its frequency value, equaled to 1328.7 cm^{-1} is slightly shifted from usually observed value near 1332 cm^{-1} . It demonstrates the implantation effect on the whole sample, indicating on renormalization of optical phonon in diamond matrix, although the thickness of the sample $\simeq 1 \text{ mm}$ is much greater, than the thickness of the near-surface ion beam modified region, which does not exceed $7 \mu\text{m}$, (implanted copper atom concentration is characterized by narrow peak with maximum at $\approx 6.5 \mu\text{m}$) [5]. Secondly, the spectrum of the same ion beam modified region is characterized by the quite different set of lines. The RS-lines with peak positions 165.3 (feature), 209.8 (feature), the lines at 354.6, 641.8, 977.1 ($\pm 1 \text{ cm}^{-1}$), $1274.1 \pm 2 \text{ cm}^{-1}$ and weakly pronounced lines at $1569 \pm 3 \text{ cm}^{-1}$, $1757 \pm 5 \text{ cm}^{-1}$ were observed in RS-spectrum, corresponding to given case. Moreover, all the lines listed are superimposed now with very broad (its linewidth value is $1720 \pm 20 \text{ cm}^{-1}$) asymmetric line with peak position $1160 \pm 10 \text{ cm}^{-1}$. The spectrum of boron implanted sample was measured in the range (1000 - 2100) cm^{-1} by excitation from implanted side. It is seen, that the spectra of boron and copper implanted samples are qualitatively similar in given spectral range. However numerical values of the peak positions are slightly different, they are - 1212.3 ± 1 , 1772.5 ± 1 , $2011 \pm 0.5 \text{ cm}^{-1}$ in the spectrum, which is characteristic of the only ion beam modified region and characteristic diamond line with peak position $1331.95 \pm 0.1 \text{ cm}^{-1}$ was also presenting. Its frequency value coincides with the value of optical phonon peak in conventional natural diamonds. The presence of diamond RS-line by excitation from implanted side can be determined by two factors. Firstly the effective thickness of ion beam modified region is substantially less ($\simeq 1.4 \mu\text{m}$), it allows to suggest, that laser excitation can reach an unimplanted region. It can also suggest, that the modification is not entire in near surface region. It is interesting, that there is regularity in peak position shift, which is increasing with frequency increase and it is equal to 2.8, 7, 11.3 cm^{-1} correspondingly. It is strong indication, that all three lines belong to the same optical system. To discuss the results of RS-studies, let us rep-

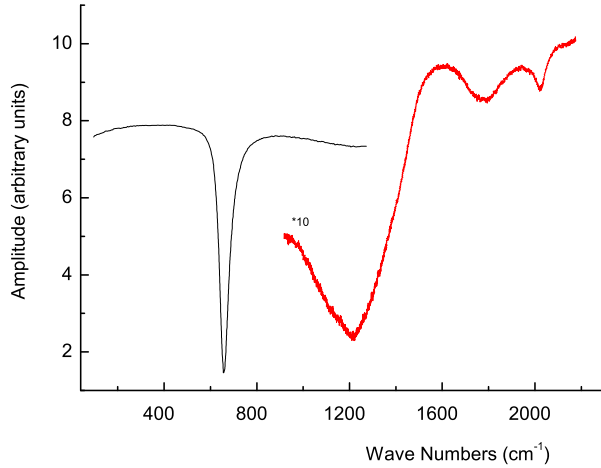


FIG. 1: Spectral distribution of Raman scattering intensity in diamond single crystal, implanted by high energy copper ions, the excitation is from implanted side of the sample.

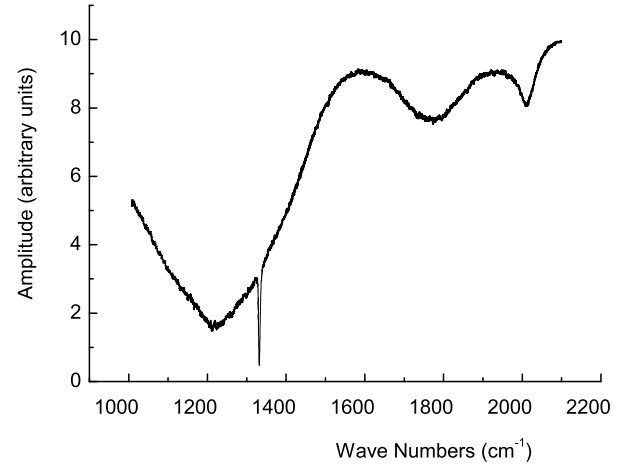


FIG. 3: Spectral distribution of Raman scattering intensity in diamond single crystal, implanted by high energy boron ions, the excitation is from implanted side of the sample.

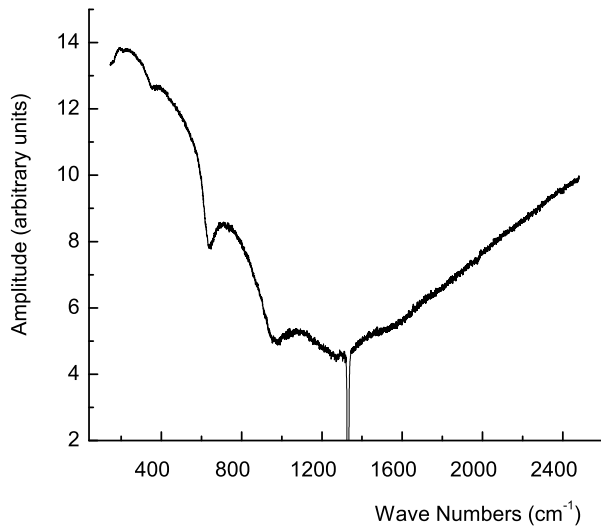


FIG. 2: Spectral distribution of Raman scattering intensity in diamond single crystal, implanted by high energy copper ions, the excitation is from unimplanted side of the sample.

resent the model of structures, produced by high energy ion implantation. A number of the studies with various methods and with the samples implanted with various ions, energy, fluences has been performed. See, for instance, review article [6] on ESR-studies, which were the most informative in structure identification. It was established the formation of quasionedimensional spatially ordered tracklike carbon structures, that is the structures with diameter of nanorange. It was in fact the discovery of new carbon phase - carbon nanotubes, incorporated

in diamond matrix, or occupying all near surface volume region. It is remarkable, that the first report is related to 1990, which was made during 1990 IBMM-Conference, Knoxville, USA and it was repeated at E-MRS 1990 Fall Meeting, Strasbourg, France, that is nothing was at that time known on the japan discovery of free NTs, related to 1991. All subsequent studies have confirmed given initial conclusion [6]. The most designing among the results obtained were the values of g -tensor components. They are in Cu -implanted sample $g_1 = 2.00255$ (it is minimal g -value and it is $g_{||}$ principal direction of axial g -tensor, at that it coincides with ion beam direction), $g_2 = g_3 = g_{\perp} = 2.00273$, the accuracy of relative g -value measurements is ± 0.00002 [5]. So we see on the one hand, that it was possible to determine anisotropy of g -values with very high precision, that indicates on the very perfect and homogeneous NTs, produced with strict axial symmetry along ion beam direction, if to take into account the origin of paramagnetic centers (PC) in ion produced NT to be paramagnetic π -solitons (SSH-solitons), considered to be mobile PC, mapping the distribution of π -electron density along whole individual chain [6], [7]. On the other hand, the g -value of paramagnetic π -solitons in trans-polyacetylene, equaled to 2.00263 [8], gets in the middle of given rather narrow interval of g -value variation of PC in ion produced NT. Although anisotropy of paramagnetic π -solitons in t-PA, which are also considered to be mobile paramagnetic centers, mapping the distribution of π -electron density along whole individual t-PA chain, is not resolved by ESR measurements directly (which in fact is the indication, that chemically produced t-PA is less perfect in comparison with NTs in diamond matrix), there are indirect evidences on axial symmetry of π -solitons in t-PA too, [9] [10]. Consequently, the value 2.00263 is mean value and it coin-

cides with accuracy 0.00002 with mean value of aforementioned principal g-tensor values of PC in NTs. Given coincidence becomes to be understandable now, if to take into account the results of [4], where is shown, that SSH-model is applicable to zigzag NT. In the frames of SSH-model zigzag NT is considered to be the system of n equivalent t-PA chains interacting between themselves. Let us give some details, concerning SSH-model. It is remarkable, that SSH-model contains in implicit form along with the physical basis for the existence of solitons, polarons, breathers, formed in π -electronic subsystem (π -solitons, π -polarons, π -breathers), also the basis for the existence of similar quasiparticles in σ -electronic subsystem, that is σ -solitons, σ -polarons, σ -breathers. The cause is the same two-fold degeneration of ground state of the whole electronic system, energy of which has the form of Coleman-Weinberg potential with two minima at the values of dimerization coordinate u_0 and $-u_0$. The shapes, for instance, of π -solitons and σ -solitons can be given by the expression with the same mathematical form

$$|\phi(n)|^2 = \frac{1}{\xi_{\pi(\sigma)}} \text{sech}^2 \left[\frac{(n - n_0)a}{\xi_{\pi(\sigma)}} - v_{\pi(\sigma)} t \right] \cos \frac{n\pi}{2}, \quad (1)$$

where n, n_0 are variable and fixed numbers of CH -unit in CH -chain, a is $C - C$ interatomic spacing projection on chain direction, $v_{\pi(\sigma)}$ is $\pi(\sigma)$ -soliton velocity, t is time, $\xi_{\pi(\sigma)}$ is $\pi(\sigma)$ coherence length. It is seen, that π -solitons and σ -solitons differs in fact the only by numerical value of coherence length. Given difference can be evaluated even without numerical calculation of the relation, which determines the shift of ground state energy of extended system by presence of localized perturbation. Actually it is sufficient to take into account the known value of ξ_π and relationships [14]

$$\xi_{0\pi} = \frac{\hbar v_F}{\Delta_{0\pi}}, \xi_{0\sigma} = \frac{\hbar v_F}{\Delta_{0\sigma}}, \quad (2)$$

where $\Delta_{0\sigma}, \Delta_{0\pi}$ are σ - and π -bandgap values at $T = 0K$, v_F is Fermi velocity. Theoretical value ξ_π in t-PA is $7a$, and it is low boundary in the range $7a - 11a$, obtained for ξ_π from experiments [15]. Taking into account the relationships (2), using the value $\frac{\Delta_\pi}{\Delta_\sigma} \approx 8.8$, which was evaluated from t-PA band structure calculation in [16], and mean experimental value of coherence length $\xi_\pi = 9a$ we obtain the value $\xi_\sigma \approx 0.125nm$. It means, that the width of space region, occupied by σ -soliton in t-PA is $\approx 0,5nm$, that is SSH- σ -solitons are much more localized in comparison with SSH- π -solitons. Similar conclusion takes place for SSH- σ -polarons representing itself the soliton-antisoliton pair, that is the width of space region, occupied by SSH- σ -polarons is evaluated to be $\approx 1nm$. SSH- σ -polarons have recently been experimentally detected in related material -carbynes [17], where the formation of polaron lattice (PL) was proposed. It was established, that two components of each elementary unit, that is, of each polaron, possess by two equal in values electrical own dipole moments, proportional to spin,

which was called electrical spin moments (ESM), with opposite directions. It was shown, that experimental results agree well with proposal of PL-formation, which means in fact the formation of antiferroelectrically ordered lattice of quasiparticles. Given lattice consists of 2 sublattices, corresponding to soliton and antisoliton components of polaron. Corresponding chain state is optically active and it is characterized by the set of lines in IR-spectra, which were assigned with new phenomenon - antiferroelectric spin wave resonance (AFESWR) Central mode is conventional antiferroelectric resonance (AFR) mode, its value $\nu_p^\sigma(C)$ in carbyne sample studied was 477 cm^{-1} . Let us remember that carbynes are organic quasionedimensional conductors with the simplest, consisting the only of the carbon atoms, chain structure. At the same time the presence of two electronic π - and π_y -subsystems, which are "hung" on σ -subsystem means that the ground electronic state is similar to twodimensional Coleman-Weinberg potential with four minima at the values of dimerization coordinate u_0 and $-u_0$. In other words, ground electronic state in carbynes is four-fold degenerate, which leads to a substantially more rich spectrum of possible quasiparticles, discussed in [18], [19].

Known value $\nu_p^\sigma(C)$ of the frequency of SSH- σ -polaron IR-active mode in carbynes allows to estimate the range for expected values $\nu_p^\sigma(t - PA)$ and $\nu_p^\sigma(NT)$ for the frequencies of SSH- σ -polaron IR-active mode in t-PA and in nanotubes of zigzag kind. Really, the known relationship for the vibration frequencies of similar centers, that is the relationship

$$\frac{\nu_p^\sigma(t - PA)}{\nu_p^\sigma(C)} = \sqrt{\frac{g(C)m_p^\sigma(C)}{g(t - PA)m_p^\sigma(t - PA)}} \quad (3)$$

is always takes place. Here $m_p^\sigma(C)$ $m_p^\sigma(t - PA)$ are SSH- σ -polaron masses, $g(C)$, $g(t - PA)$ are degeneration degree factors in carbynes and in t-PA respectively. It is evident, that the ratio of polaron masses can be replaced by the soliton mass ratio, the expression for which is [12], [15]

$$m_s = \frac{4}{3\epsilon} \left(\frac{u_0}{a} \right)^2 M, \quad (4)$$

where M is mass of $C - H$ unit and carbon atom in the case of t-PA and carbyne respectively. Then, taking into account the relationships (2) we obtain $\nu_p^\sigma(t - PA) \in (487.2, 759.7) \text{ cm}^{-1}$ and $\nu_p^\sigma(NT) \in (507.1, 790.7) \text{ cm}^{-1}$. The theoretical values for carbyne σ -bandgap from calculation in [20] were used. They are strongly dependent on the number of carbon atoms in elementary units of carbyne chain, that determines the relatively large spectral regions above indicated. Let us compare the expected value for IR-active SSH- σ -polaron mode in t-PA with experimental data. The mode with the frequency near 540 cm^{-1} , which was observed in infrared photoinduced spectra of t-PA gets to interval $(487.2, 759.7) \text{ cm}^{-1}$ and can represent itself AFR mode in σ -polaron lattice. It means, that earlier ascribing IR-line near 540 cm^{-1} to

Goldstone SSH- π -soliton mode has to be reinterpreted. Given conclusion is confirmed additionally by the following arguments. 1. The frequency value in $\sim 540 \text{ cm}^{-1}$ is too large for any Goldstone mode. For comparison, so called "breathing" modes in related materials (see also further consideration) - carbon single wall NTs - were observed in the range between $140\text{-}200 \text{ cm}^{-1}$ [21], moreover shear modes in nested NTs have the frequency values at $49, 58 \text{ cm}^{-1}$ [22]. Similar values of the frequency seems to be reasonable to expect for Goldstone mode of SSH- π -solitons. It is remarkable, that at the same time the lines at $\sim 700 \text{ cm}^{-1}$ [22] in nested NTs and in the range $750\text{-}790 \text{ cm}^{-1}$ for single wall NTs [21] were also presenting. Naturally, authors of [21], [22] do not refer given modes to Goldstone modes, although their frequency values are intermediate between $\sim 540 \text{ cm}^{-1}$ and $\sim 900 \text{ cm}^{-1}$, which were considered to be Goldstone modes in undoped and doped t-PA respectively. It is evident, that the pinning role in undoped NT and undoped t-PA is comparable. 2. The resemblance in the set of lines, which belong to the same absorbing center and which are in given case the result of fine (vibronic) splitting of the same electronic level has to be revealed. The resemblance is concerned first of all the line shapes and linewidths. Comparison of line shapes and linewidths of the line $\sim 540 \text{ cm}^{-1}$ and the second line with higher frequency value in Fig.39 [15] shows, that the differences in line shapes and linewidths are very large. It means, that compared lines cannot belong to the same center. 3. Both the lines $\sim 540 \text{ cm}^{-1}$ and $\sim 900 \text{ cm}^{-1}$ are strongly intensive. It testifies to the favour of proposal, that $\sim 540 \text{ cm}^{-1}$ absorption line in photoexcited t-PA is analogue of 477 cm^{-1} carbyne line, that is, it can really be assigned with AFR mode in σ -polaron lattice, produced in t-PA by photoexcitation along with charged π -soliton formation. We also wish to remark, that the line $\sim 900 \text{ cm}^{-1}$ can also belong to σ -polarons in another charge state and that AFSWR can in principle be observable by suitable geometry of experiment in both the cases.

Taking into account the calculation of the frequencies of vibration active modes for NTs of various kind in [22], we can conclude a priori, without additional calculations, that in spectral interval, which is near to $(507.1, 790.7) \text{ cm}^{-1}$ σ -polaron in zigzag NT will have the vibration active mode, which is active in Raman scattering. Really, on the one hand, the formation of quasiparticles in zigzag NT of the same type, that those ones in t-PA, was proved in [23]. On the other hand, the calculation in [22] indicates on the presence of even and uneven modes in above indicated interval with frequencies, which have close values, at that some of even Raman active modes are dependent on NT diameter value. Given calculation does not take into consideration the soliton and polaron formation. However σ -polaron or σ -soliton formation does not violate the symmetry of task, that means, that for zigzag NT to the IR σ -polaron mode in range $(507.1, 790.7) \text{ cm}^{-1}$ will correspond the Raman σ -polaron mode at close frequency range. Therefore we come to conclusion, that

intensive line $656.8 \pm 0.2 \text{ cm}^{-1}$ in RS-spectrum, presented in Fig.1 can be assigned with AFR mode of σ -polaron lattice, produced in NTs, while the lines, $1215 \pm 1 \text{ cm}^{-1}$, $1779.5 \pm 1 \text{ cm}^{-1}$ and $2022.3 \pm 0.5 \text{ cm}^{-1}$ is revival part [2] in its frequency representation [4] of Rabi wave packet, which is produced in result of interaction of σ -polaron lattice with quantized EM-field. Given identification is confirmed by the following. It is well known, that is in the case of point absorbing centers Rabi frequency is linearly dependent on the amplitude of oscillating EM-field. To lesser value of exiting amplitude correspond also lesser values of the frequencies of traveling Rabi waves. Given tendency takes also place for high frequency components of wave packets, that follows from the analysis of Fourier transform of temporal dependence of the integral inversion for a coherent initial state of light, given in [2]. Further, it is evident, that amplitude of laser wave, penetrating in ion beam modified region, is less by excitation from unimplanted side in consequence of some absorption in the unimplanted volume of diamond single crystal. We see, that really, the experimental frequency values $1569 \pm 3 \text{ cm}^{-1}$, $1757 \pm 5 \text{ cm}^{-1}$ of two high frequency components are substantially lesser in given case, than $1779.5 \pm 1 \text{ cm}^{-1}$ and $2022.3 \pm 0.5 \text{ cm}^{-1}$, observed by excitation from implanted side, compare Fig.2 and Fig.1. The AFR mode is observed now at $641.8 \pm 1 \text{ cm}^{-1}$ instead of $656.8 \pm 0.2 \text{ cm}^{-1}$, which indicates on the presence of hysteresis relatively the direction of excitation wave propagation. The lines at $354.6, 977.1 (\pm 1 \text{ cm}^{-1})$ seem to be assigned with two AFSWR modes, that is, there is splitting of AFR-mode into AFSWR spectrum. The substantial decrease of relative intensity of $641.8 \pm 1 \text{ cm}^{-1}$ mode in comparison with $656.8 \pm 0.2 \text{ cm}^{-1}$ testifies in favour of given assignment. It is seen, that AFSWR-splitting is rather large and it is comparable with the splitting between two polaron vibration levels. It means, that linear AFSWR-theory, developed in [17], which predicts a set of equidistant AFSWR-modes, arranged the left and the right of central mode, will be not applicable for given case. Really AFSWR-modes are not equidistant, they are shifted on distances $335.3, 287.2 \text{ cm}^{-1}$ from main AFSWR-mode. At the same time mean value of AFSWR-splitting is 311.3 cm^{-1} and it is close to two-fold value of AFSWR splitting (which is equal to 150 cm^{-1} assigned with σ -polaron lattice in carbynes [17]. Given approximately two-fold splitting increase seems to be result of corresponding increase of spin wave resonance splitting, registered by Raman spectroscopy methods in comparison with SWR splitting, registered by IR spectroscopy methods, predicted in [25] and observed earlier by ferroelectric SWR study in carbynes [26]. To explain so significant differences in the physics of processes by the change of the direction of the excitation wave propagation, we have to take into consideration the following. Any SWR-excitation in 1D-systems is strongly dependent on the geometry of experiment, that is, on the mutual orientation of chain axis and vectors of intracrystal electric field, which determines the splitting of energetic

levels, and electrical component of EM-field, calling the spectroscopic transitions like to possibility of the excitation of ferromagnetic spin wave resonance in carbynes [27]. NT axes in zigzag NTs are not linear in the end of ion run and they are generatrices of the figure of onion-like shape, that provides for necessary geometry for AF-SWR excitation in photon dressed state. Moreover the appearance of very broad line seems to be indication on the excitation by given laser wave propagation direction

of the movement of polaron lattice itself. Therefore NTs represent themselves the example of the system, which strongly interact with EM-field. Experimental detection of Rabi wave packets confirms the theory, developed in [2] on the one hand. On the other hand it means, that semiclassical description of spectroscopic transitions in NTs and in the systems like them cannot be appropriate, which seems to be substantially raising the practical concernment of QED-theory.

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